SWIFT HEAVY ION IRRADIATION EFFECTS ON He AGGLOMERATION IN ZrN AND TiZrN CERAMICS

Arno Janse van Vuuren[1)], Vladimir Skuratov[2)], Vladimir Uglov[3)], Alexander Sohatsky[2)]

[1)]*Centre for HRTEM, University Way, Summerstrand, Port Elizabeth, South Africa, arnojvv@gmail.com*

[2)]*Flerov Laboratory for Nuclear Research, Joint Institute for Nuclear Research, Dubna, Russia, skuratov@jinr.ru, sohatsky@jinr.ru*

[3)]*Belarusian State University, Minsk, Belarus, uglov@bsu.by*

The development of novel materials with future nuclear applications is of ongoing concern.As part of this investigation we address two major issues concerning such applications. Firstly, improving the performance of structural materials in the reactor core for new and existing reactors and also the processing and storage of nuclear waste. Nanocrystalline-ZrN and -TiZrN have been identified as candidate materials for inert matrix fuel hosts and as a coatings for structural materials respectively. These materials will experience high levels of different types of radiation in the reactor core such as fission fragments and α-radiation. The effects of different types of radiation on the long term stability of these materials however still needs to be determined. The effects of fission fragments are simulated by means of swift heavy ion implantation and α-radiation by He implantation. The combined effects of SHIs and He on these materials is also studied, since recent studies have indicated that SHI irradiation may significantly modulate hydrogen and helium behaviour in materials. ZrN and TiZrN samples were implanted with 30 keV He and 167 MeV Xe and in some cases also 695 MeV Bi, the samples were subsequently annealed to temperatures between 600 and 1000 °C for 20 minutes. The microstructure of the annealed samples was investigated by scanning electron microscopy (SEM) and transmission electron microscopy (TEM) before and after SHI implantation. Results indicated that post irradiation heat treatment induces exfoliation at a depth that corresponds to the end-of-range of 30 keV He ions, however the formation of blisters was suppressed after irradiation with SHIs. It is also important to note that latent ion tracks were not observed in any of the samples.

Introduction

Recent studies have shown that swift heavy ion irradiation may significantly modulate hydrogen and helium behaviour in certain materials [1, 2]. This phenomenon is of considerable practical interest for various ceramics and semiconductors, specifically for candidate materials for use as inert matrix (IMs) fuel hosts and for coatings for structural materials in the nuclear reactor.

Inert matrix fuel hosts have been suggested as a means of processing transuranic waste products resulting from the nuclear fuel cycle andspecialised coatings for structural components in the nuclear reactor to improve corrosion resistance.

These materials accumulate helium via (n,α) reactions and will also be subjected to irradiation by high energy fission fragments in the nuclear reactor environment [3]. The inherent properties of ZrN and TiZrN ceramics have led to their identification as a candidate materials for use as an inert matrix fuel hosts [4] and coatings for structural materials respectively. Most of the minor actinides and plutonium are accommodated in the ZrN lattice. TiZrN is compatible with the current zircaloy fuel tubes used in many reactors. The adhesion, radiation stability, corrosion resistance and radiation resistance of the TiZrN-Zircaloy system however still needs to be confirmed.

Certain studies have also indicated that some nanocrystalline (nc) materials have improved radiation tolerance compared to their micro- and single-crystalline counterparts [5, 6]. This suggests that nc-ZrN and nc-TiZrN could possibly have superior radiation tolerance.

Experimental

The ZrN and TiZrN layers used in this investigation were produced via vacuum arc-vapor deposition. The layers are approximately 20 μm (ZrN) and 4 μm (TiZrN) in thickness.

In this study low energy He ions were used to simulate the effects of α-particles. High energy Xe and Bi ions were used to simulate the effects of fission fragments in nc-ZrN and nc-TiZrN. The combined effects of low and high energy radiation were also studied, since these materials will be subjected to both types of radiation in the nuclear reactor core.

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) techniques were used to study the various samples. The ZrN and TiZrN layers were irradiated with 30 keV He and subsequently with 167 MeV Xe. In order to determine the effects of increasing stopping power some ZrN samples were also irradiated with 695 MeV Bi ions subsequent to the He irradiation. All samples were annealed at temperatures between 600 and 1000 °C for 20 minutes in an inert argon atmosphere.

TEM lamellae were produced by means of a FEI Helios Nanolab FIB-SEM, which was also used for SEM analysis. The lamellae were investigated with a JEOL 2100 LaB_6 TEM operated at 200 kV.

Results and Discussion

TEM analysis confirmed the nanocrystalline morphologies of both the ZrN and TiZrN layers. In most sample the crystal grain are elongated perpendicular to the surface but much narrower in the lateral direction. TEM analysis also revealed that post irradiation heat treatment induces blistering at a depth that corresponds to the end-of-range of 30 keV He ions as shown in Fig. 1 (ZrN) and Fig. 2 (TiZrN). Blisters were observed in nc-ZrN at all He fluences used in this investigation i.e. 1×10^{16}, 1.5×10^{16} and 5×10^{16} cm^{-2}. The size and number of blisters and bubbles in nc-ZrN increased with He fluence as expected. The TiZrN layers were only irradiated with He ions to a fluence of 5×10^{16} cm^{-2}.

Analysis of He/Xe irradiated samples revealed that the electronic excitation effects resulting from

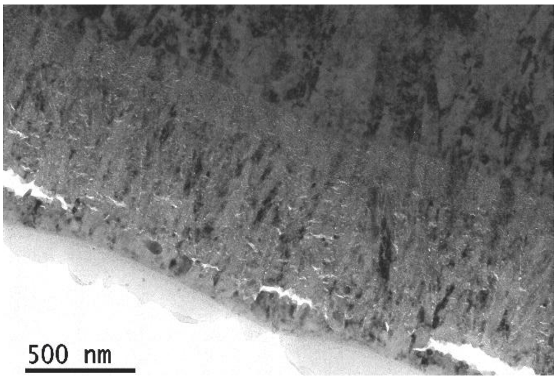

Fig. 1. A bright field TEM micrograph showing a cross-sectional view of a nanocrystalline ZrN layer irradiated with He to a fluence of 1×10^{16} cm^{-2} and annealed at 700°C.

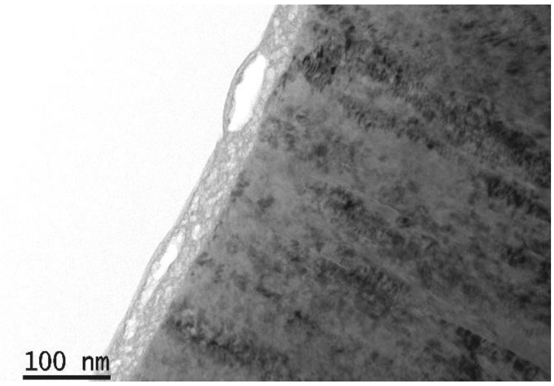

Fig. 2. A Bright field TEM micrograph showing a cross-sectional view of a nanocrystalline TiZrN layer irradiated with He to a fluence of 5×10^{16} cm^{-2} and annealed at 700 °C.

high energy Xe ions supress the formation of helium blisters. This can clearly be seen in Fig. 3 (ZrN) and Fig. 4 (TiZrN) where the amount of blistering and bubble formation in the layers is significantly less than in the sample irradiated with He only.

The suppression of blister formation is less efficient at higher He fluences, but is still evident. He/Bi samples however do not show the same effects which suggest that some threshold fluence value may be required for blister suppression, since the maximum Bi fluence is lower than that of Xe. This also suggest that the suppression of He agglomeration due to SHIs is possibly independent on the electronic energy loss S_e, however further investigation is required in order to confirm this.

Conclusion

The results of this investigation suggest that He irradiated nanocrystalline ZrN is prone to the formation of He gas bubbles/blisters. The formation of these blisters may ultimately lead to material failure,

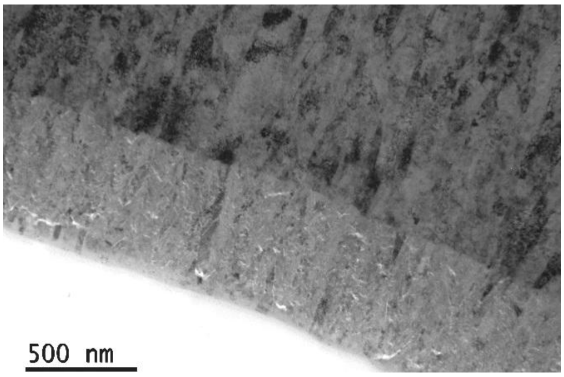

Fig. 3. A bright field TEM micrograph showing a cross-sectional view of a nanocrystalline ZrN layer irradiated with He and Xe to fluences of 1×10^{16} cm^{-2} and 8×10^{13} cm^{-2} respectively.

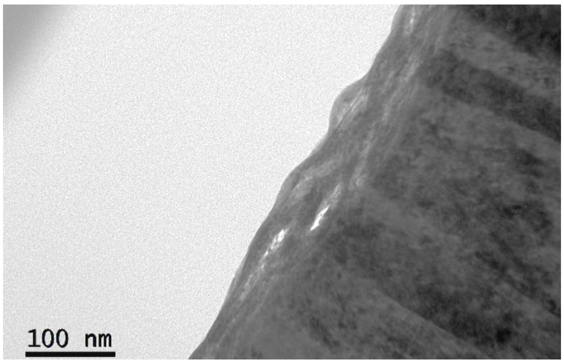

Fig. 4. A bright field TEM micrograph showing a cross-sectional view of a nanocrystalline TiZrN layer irradiated with He and Xe to fluences of 5×10^{16} cm^{-2} and 8×10^{13} cm^{-2} respectively.

which will be of great concern for materials with nuclear applications. These effects may however be mitigated by the electronic excitation effects from certain high energy heavy ions such as Fission Fragments.

Acknowledgments

The work is sponsored in part by the NRF of South Africa and BRFBR-JINR program, project F15-006.

References

1. *Reutov V.F. et al.* // Tekhn. Phys. 2009. V. 79. P. 63.
2. *Li B.S., Zhang C.H. and Yang Y.T.* // Rad. Eff. and Defects in Solids. 2012. V. 167. P. 212.
3. *Lee Y.-W. et al.* // Metals and Materials International. 2001. V. 7. P. 159.
4. *Kleykamp H.* // J. Nucl. Mater. 1999. V. 275. P. 1.
5. *Nita, N. et al.* // Phil. Mag. 2005. V. 85. P. 723.
6. *Janse van Vuuren, A. et al.* // J. Nucl. Mater. 2013. V. 442. P. 507.